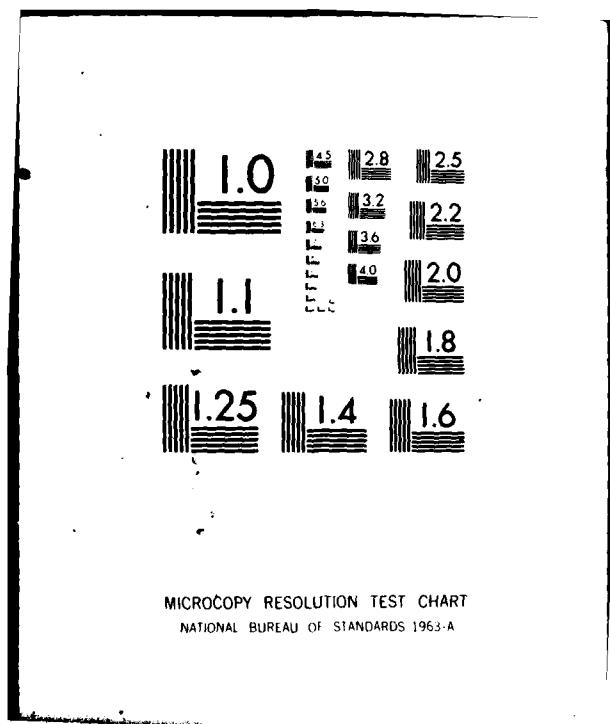


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TECHNICAL REPORT NO. 6

Surface Magnetic Field/Laser Synergistic
Effects on the Predissociation of Adsorbed Diatomic
Molecules

by

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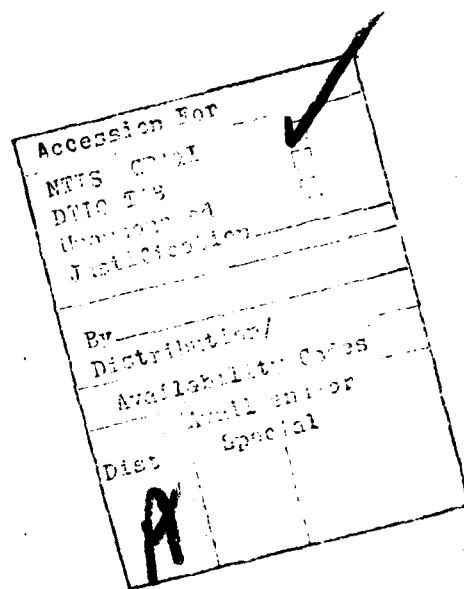
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20. Abstract (continued)

are discussed along with their dependence on laser power densities.



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Surface Magnetic Field/Laser Synergistic
Effects on the Predissoociation of Adsorbed Diatomic
Molecules^{a)}

Dilip K. Bhattacharyya, Kai-Shue Lam and
b)
Thomas F. George

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Abstract

Synergistic effects of the surface magnetic field (SMF) and the laser on the predissociation of adsorbed diatomic molecules are studied theoretically. It is observed that the couplings between the laser-dressed and SMF-split molecular potentials create interference effects within the nuclear motion. These effects can cause enhancement or diminution in predissociation rates as compared to the analogous gas-phase laser-induced process. SMF/laser-induced predissociation rates for a few specific examples are derived using the semiclassical S-matrix formalism, and results for the enhancement and diminution are discussed along with their dependence on laser power densities.

I. Introduction

Recently, the effect of the surface magnetic field (SMF), most notably from ferromagnetic metallic surfaces, has been discussed¹ in relation to the dissociation of adsorbed diatomic molecules. This field (often referred to as the exchange or Weiss field), originates from the exchange interaction between unpaired electronic spins on the metallic surface, and for the case of iron, reaches a magnitude of $\sim 10^7$ G.² In the presence of the SMF, multiplet molecular terms of the adsorbed molecule will undergo Zeeman splitting, giving rise to multiple branches and configurations where dissociative and bound channels which normally do not cross may do so. These crossing channels are then coupled through the interaction of the diatomic electronic spin and the SMF. As a result, there is a finite probability for the higher vibrational levels to predissociate by virtue of their possessing extra kinetic energy or by tunneling. An example is the H_2/Ni system, where the SMF of the metal splits the first excited $^3\Sigma_u^+$ potential curve into three branches and the lowest branch crosses the ground $^1\Sigma_g^+$ curve (see discussion in Section III, Fig. 1, and Reference 1.)

From the situation described above (also applicable to O_2 and N_2), it becomes obvious that in the presence of the surface, only the higher vibrational levels can predissociate. The low-lying levels, however, which are in fact the most populated, cannot decay by this mechanism. Moreover, for molecules such as NO

and CO, where the ground and the first excited potential surfaces have asymptotic energy gaps larger than the SMF splittings, the SMF cannot create any crossing between the ground and excited split branches. As a result, no vibrational level predissociates for such molecules.

To obtain dissociation of molecules like NO and CO, or of the low-lying levels of H_2 , N_2 etc., a laser in addition to the surface may prove effective. The laser serves to supply the extra energy to bring non-crossing dissociative and bound surfaces into crossing configurations, and in the cases where they already cross, to create more crossings. In a previous letter,³ we have discussed such a combined SMF/laser synergistic effect on the dissociation of low-lying states of H_2 . It was found that for these states, the SMF/laser-induced predissociation rate is more pronounced than the gas-phase laser-induced predissociation, whereas in the presence of the surface alone these states have a negligible predissociation rate.

In this paper we shall extend the treatment introduced in Reference 3 and apply it to a few more specific cases (having different multiplicities for the ground and excited electronic curves) with the intention of studying the relative effectiveness of the laser on different systems. Our investigation will be qualitative, in that the detailed physical characteristics of the admolecule-surface system will be ignored. The only crucial features of the system that we will exploit are the qualitative presence of the

splitting and the dissociative-bound channel couplings. In most of the cases studied, enhancement is predicted for all laser powers. In one case (O_2), however, diminution is also possible, with enhancement favored only at high laser powers. In Section II we present a theoretical formalism for the treatment of the laser-induced predissociation of an adsorbed molecule based on the electronic-field representation⁴ and the semiclassical S-matrix formalism.⁵ In Section III, formulas for predissociation rates of the different specific cases are stated. Finally, in Section IV, results demonstrating the effect of the laser on admolecule predissociation are presented, and their physical significance discussed. Estimates of the required laser powers for the attainment of reasonable enhancement are also given.

II. Theory

A convenient mode of description for the radiative interaction between the molecular electronic states, some of which may be split by the SMF, is the electronic-field representation.⁴ In this representation, the basis states are direct product states of the molecular system (in our case the adsorbed molecule as modified by surface effects) and the radiation field [which is usually described in the Fock (number-state) representation]. Employing the rotating-wave approximation,⁶ one can then limit the consideration of absorption and emission to single-photon processes; and radiative coupling is present only between states whose photon numbers differ by one.⁶ [This coupling can either be of the electric-multipole or magnetic-multipole type, depending on the symmetry characters of the molecular states involved]. Molecular states which do not cross in the field-free configuration may, when the field is present, generate real crossings (in the diabatic representation) or avoided crossings (in the adiabatic representation) - see Fig. 1. [We should mention that the detailed structure of the potential surfaces of the adsorbed species may of course deviate considerably from the corresponding gas-phase structure; but this will in no way affect our present qualitative investigation of the laser-induced effects.]

The avoided crossings generate additional bound wells for vibrational motion of the adsorbed species (see the discussion of specific examples in the next section and Fig. 1). However, since

all of these wells involve crossings or avoided crossings, no motion in them is truly bound; and the pseudo-bound states may be described as having shifts and widths for their energy levels. These are induced by a certain probability of 'leakage' away from motion in a particular well, most conveniently visualized, semiclassically, as taking place around the avoided crossings. From the widths of the energy levels in the various bound wells, one can readily obtain the predissociation rate of the adsorbed molecular system.

The interference of pseudo-bound motion in a multi-well system is readily described within the framework of the semiclassical S-matrix formalism.⁵ The bound-state energies, as well as the shifts and widths, can be directly obtained by locating the poles of the S-matrix element for oscillatory motion within the wells. The rules for the construction of the semiclassical S-matrix for oscillatory motion are summarized as follows: (i) There is a factor of $\exp(2ia)$ for every round-trip within a well, a = action integral for a single pass; (ii) For every turning-point encounter, there is a factor of $\exp(-i\pi/2)$; (iii) For every crossing over at an avoided crossing, there is a factor of \sqrt{p} , p = local transition probability at an avoided crossing; and (iv) For every non-crossing over at an avoided crossing, there is a factor of $\sqrt{1-p}$. The factor p may be estimated by using the Landau-Zener form⁷ or computed by using, for instance, the Miller-George semiclassical treatment.^{4,8,9} For a combination of N wells with various configurations

of avoided crossings, then, the S-matrix can be expressed as:

$$S_N \propto \sum_{n_1=0}^{\infty} e^{(2i\alpha_1 - i\pi)n_1} (w_1)^{n_1} \sum_{n_2=0}^{\infty} e^{(2i\alpha_2 - i\pi)n_2} (w_2)^{n_2} \dots \sum_{n_N=0}^{\infty} e^{(2i\alpha_N - i\pi)n_N} (w_N)^{n_N}. \quad (1)$$

In Eq. (1) α_i is the action integral for a single pass in the i -th well, $w_i > 0$ is the combination of p and $(1-p)$ factors in the i -th well according to rules (iii) and (iv) above, and each term in the N -fold sum represents a trajectory which makes a round-trip in the first well n_1 times, the second well n_2 times, etc. The sum in Eq. (1) can easily be carried out to obtain the proportionality

$$S_N \propto [(1+w_1 e^{2i\alpha_1}) (1+w_2 e^{2i\alpha_2}) \dots (1+w_N e^{2i\alpha_N})]^{-1}. \quad (2)$$

From Eq. (2) it is seen that the poles for S_N are obtained by setting

$$1 + w_i \exp(2i\alpha_i) = 0. \quad (3)$$

The action integrals α_i are then given by

$$\alpha_i = (n + \frac{1}{2})\pi - \frac{i}{2} \log \left(\frac{1}{w_i} \right), n=0,1,2,\dots \quad (4)$$

Comparing with the expansion

$$\begin{aligned} \alpha_i(E) &\approx \alpha_i(E_0) + \alpha'_i(E_0)(E-E_0) \\ &\equiv \alpha_{0i} + \alpha'_{0i}(E-E_0) \end{aligned} \quad (5)$$

where E_0 is the exact bound-energy in the i -th well, and observing that $\alpha_{0i} = (n + \frac{1}{2})\pi, n=0,1,2,\dots$ by the WKB condition and $\alpha'_{0i} \approx \pi/hv_i = \text{constant}$ (v_i = exact resonance frequency in the i -th

well), we obtain for the width Γ_i , for all pseudo-bound levels in the i -th well,

$$\Gamma_i = \frac{1}{\alpha_{0i}} \log \left(\frac{1}{w_i} \right), \quad (6)$$

if we put $E \approx E_0 - \frac{1}{2} \Gamma_i$. [To our approximation, the shift (ΔE) is zero.]

The total predissociation rate Γ , however, is not given by the simple sum of the Γ_i , since there are finite (and different) probabilities for the molecular system to 'live' in the different wells, depending on the local transition probabilities p . It follows that the statistical weights to be assigned to the Γ_i for their contributions to Γ must be different. Now any configuration of N wells can always be divided into m groups ($m < N$) such that the members in each group all have a certain 'segment' of the potential surfaces (in the electronic-field representation) in common, and this 'segment' always cuts a turning point (see Section III). The number of such groups is, in fact, equal to the multiplicity of the ground-electronic state in question, and the relative probability of the system 'living' in each group can be taken to be $1/m$. Moreover it can be easily verified that, for each group of wells,

$$\sum_{i \in \text{group}} w_i = 1. \quad (7)$$

Hence it is reasonable to assume that the statistical weight to be assigned to each Γ_i should be w_i/m . [From (7), $\sum_i (w_i/m) = 1$, the summation index here is over all wells.] We then have the final

expression for the total predissociation rate Γ :

$$\Gamma = - \frac{1}{m} \sum_{i=1}^N \frac{w_i}{\alpha_{oi}} \log w_i . \quad (8)$$

III. Examples

We will now discuss some specific examples of adsorption of diatomic molecules on the metal surface. In all of these cases, the molecular terms other than singlet are split into multiple branches due to the SMF. The laser-dressed and the SMF-split electronic levels cross, creating interference within the nuclear motion in the wells.³ Since the splitting of the electronic levels, characteristics of the quasi-bound wells, and hence the interference within the nuclear motion, depend on the multiplicities of the electronic states involved, we will take specific examples and discuss them in separate subsections. The formulas derived in the following will be based on the situation where the laser frequency is such that all the quasibound wells are accessed.

(a) $H_2, N_2, CO/Co, Ni$ etc.

Each of these molecules has a singlet ground state and a triplet first excited state. For H_2 and N_2 , the SMF splitting may cause the lowest triplet branch to cross the ground singlet, thus inducing a minimal finite probability for predissociation of very high vibrational levels. The laser can enhance that probability by creating more crossings and channels for predissociation. For CO, the asymptotic energy gap between the ground ($^1\Sigma^+$) and the first-excited ($^3\Pi$) state is larger than the amount of splitting caused by the SMF. Hence, none of the SMF-split triplet branches crosses the ground singlet. Only a laser, in this case,

can give rise to crossing between the dressed singlet and the surface-split triplet branches.

The situation is schematically shown in Fig. 1(a).

Here the low-lying dressed singlet of H_2 or N_2 crosses the SMF-split triplet branches. Because of the different spin multiplicities of the crossing states, the laser can induce only weak couplings - spin-electric dipole (SE1) and spin-magnetic quadrupole (SM2).¹⁰ Describing the situation in the electronic-field representation, the upper three avoided crossings can be recognized as laser-field generated couplings, and the bottom crossing as the one caused by the SMF. The situation is almost similar for CO though the excited triplet is bound. Here too, the split triplets cross the dressed singlet giving rise to similar avoided crossings and predissociation channels. But, here we do not have any crossing created by the SMF. In any of these situations, the low-lying vibrational state under consideration can now dissociate through all the possible channels when the laser field is applied. Because of the singlet ground state, we only have one group of wells ($m=1$), the total number of wells being four. We can call these four wells $V_{1,1}$, $V_{1,2}$, $V_{1,3}$ and $V_{1,4}$ (Fig. 1a). $V_{1,1}$ is the diabatic well for the diabatic surface $1'$ and V_{ij} is the "adiabatic" well generated by the segments of diabatic surfaces i and j . All of these four wells contain segments of the diabatic surface $1'$. One can easily visualize these wells as one starts from the right-hand turning point. For example, the well

$v_{1'2'}$ is first generated by a segment of the diabatic surface 1', and then by a segment of the surface 2 beyond an avoided crossing (with local transition probability p_1 , in this case).

As discussed in Section II, the total width Γ (and hence the predissociation rate) can be derived from the poles of the S-matrix for oscillatory motion in the wells. After identifying the wells, we can use Eq. (8), and write for this case,

$$\Gamma = - \sum_{i=1}^4 \frac{w_i}{\alpha'_{0i}} \log w_i \quad (9)$$

Here, we have

$$\begin{aligned} w_1 &= p_1 p_2 p_3 \\ w_2 &= (1-p_1) p_2 p_3 \\ w_3 &= (1-p_2) p_3 \\ \text{and } w_4 &= (1-p_3) \end{aligned} \quad (10)$$

The four w_i 's and α'_{0i} 's have a one-to-one correspondence with the four wells. They are related as follows:

$$w_1, \alpha'_{01} \leftrightarrow v_{1'} \quad w_2, \alpha'_{02} \leftrightarrow v_{1'2'}$$

$$w_3, \alpha'_{03} \leftrightarrow v_{1'3'} \quad w_4, \alpha'_{04} \leftrightarrow v_{1'4'}$$

It is obvious from the above expressions that the w_i factors for these wells add up to 1 as required by Eq. (7).

(b) NO/Ni,Co etc.

The situation is schematically shown in Fig. 1(b). Here, both the ground and the first-excited states are doublets. The

SMF-split and laser-dressed ground doublets now cross the SMF-split doublet branches of the excited potential surface and thus create four avoided crossings. All these crossings are due to the laser alone; and, as in CO, there is no crossing created by the SMF. Since the spin is conserved, an electric-dipole transition is allowed and one expects a higher predissociation rate than for the cases discussed in Subsection III(a). Because of the doublet ground state, we have two groups ($m=2$) of wells (total of nine; six in one and three in the other) which individually satisfy Eq. (7). All the six wells in the first group contain a common segment of the diabatic surface $1'$ and those of the second group contain a common segment of the diabatic surface $2'$. Using the notation introduced in the previous subsection, we label the two groups of wells by:

Group I wells: $v_{1,1}, v_{1,32}, v_{1,3}, v_{1,42}, v_{1,42,3}$, and $v_{1,4}$;

Group II wells: $v_{2,1}, v_{2,3}$ and $v_{2,4}$.

The expression for Γ now becomes:

$$\Gamma = -\frac{1}{2} \sum_{i=1}^9 \frac{w_i}{\alpha'_{0i}} \log w_i . \quad (11)$$

We relate the w_i 's and α'_{0i} 's to the nine wells in similar order as given above. Thus, we have

$$\begin{aligned} w_1 &= p_1 p_2 \\ w_2 &= p_1 (1-p_2) (1-p_4) \\ w_3 &= p_1 (1-p_2) p_4 \\ w_4 &= (1-p_1) (1-p_3) p_4 \end{aligned}$$

$$\begin{aligned}w_5 &= (1-p_1)(1-p_3)(1-p_4) \\w_6 &= (1-p_1)p_3 \\w_7 &= p_3p_4 \\w_8 &= p_3(1-p_4) \\w_9 &= (1-p_3)\end{aligned}\tag{12}$$

It is obvious from the above equation that

$$\sum_{i=1}^6 w_i = \sum_{i=7}^9 w_i = \frac{1}{2} \sum_{i=1}^9 w_i = 1 .\tag{13}$$

(c) O₂/Co,Ni etc.

As shown schematically in Fig. 1(c), the SMF-split and laser-dressed triplet branches of the ground state, in this case, cross the unsplit excited singlet surface; and three avoided crossings are created. The laser-induced couplings are now S1 and S2. Proceeding as before, we label the three groups ($m=3$) of wells (a total nine) having the 1', 2', and 3' surfaces respectively as common segments as follows:

$$\begin{aligned}\text{Group I: } &v_{11}, v_{1'42}, v_{1'43}, v_{1'4} ; \\ \text{Group II: } &v_{21}, v_{2'43}, v_{2'4} ; \\ \text{Group III: } &v_{31}, v_{3'4} .\end{aligned}$$

For Γ , we get,

$$\Gamma = -\frac{1}{3} \sum_{i=1}^9 \frac{w_i}{\alpha'_{0i}} \log w_i ,\tag{14}$$

and relating w_i 's and α'_{0i} 's to the wells in the order given above we have,

$$\begin{aligned}w_1 &= p_3 \\w_2 &= (1-p_3)(1-p_2) \\w_3 &= (1-p_3)p_2(1-p_1) \\w_4 &= (1-p_3)p_2p_1 \\w_5 &= p_2 \\w_6 &= (1-p_2)(1-p_1) \\w_7 &= (1-p_2)p_1 \\w_8 &= p_1 \\w_9 &= (1-p_1)\end{aligned}\tag{15}$$

The above expression satisfies the condition

$$\sum_{i=1}^4 w_i = \sum_{i=5}^7 w_i = \sum_{i=8}^9 w_i = \frac{1}{3} \sum_{i=1}^9 w_i = 1 .\tag{16}$$

IV. Results and Discussion

Here we present qualitative results for Γ , using Eqs. (8)-(12), (14) and (15), for the three typical well-configurations discussed in section III. Our results will be compared with those for gas-phase laser-induced predissociation of the respective systems. In obtaining these results the following assumptions have been made: (i) In each case, the local transition probabilities p_i are taken to be identical, i.e., $p_i \approx p = \exp(-\theta)$, where θ is a dynamical factor dependent on the strength of the radiative coupling and the geometry of the crossing curves, and can be taken to be proportional to I , the laser power density; (ii) The derivatives of the action integrals, α'_{0i} , are taken to be the same for every well, i.e., $\alpha'_{0i} \approx \alpha'_0 = \text{const}$. These assumptions have been made purely in the interest of simplicity but are not expected to weaken our overall qualitative interpretation of the results - enhancement for the majority of the cases studied. Fig. 2 presents results for the dissociation rates in the form of $\alpha'_0 \Gamma$ vs θ , where the lower curves represent gas-phase laser-induced predissociation and the upper ones are for the adsorbed species subjected to the combined action of the SMF and the laser. The gas-phase situations, for all the cases discussed in Section III, only involve a "single avoided-crossing" configuration, and

$$\alpha'_0 \Gamma (\text{gas-phase}) = -p' \log p' - (1-p') \log(1-p') . \quad (17)$$

Because of the degeneracy of the molecular states, $p'(^1\Sigma_g^+ \leftrightarrow ^3\Sigma_u^+)$ $\sim p'(^3\Sigma_g^- \leftrightarrow ^1\Delta_g)$ $\sim \exp(-3\theta)$ and $p'(^2\Pi \leftrightarrow ^2\Sigma^+)$ $\sim \exp(-2\theta)$, if p is

taken to be $\exp(-\theta)$ for the respective cases involving adsorbed species.

For most of the adsorbed systems studied, an enhancement in the predissociation rate is observed. The O_2 case, however, shows diminution in the low laser-power region. The enhancement suggests possible synergistic effects of the surface and the laser: predissociation, for low-lying vibrational states is negligible in the presence of the surface alone, but may be more pronounced than laser-induced gas-phase predissociation when the surface and the laser are both present. This enhancement or diminution may be attributed to the interference within the nuclear motion in the extra wells generated by the SMF-split potentials.

For realistic values of I , the region $0 < \theta < 1$ will be of most interest. Since the electric dipole coupling (as in NO) is much stronger ($\sim 10^2$ - 10^3 times) than the spin-dependent couplings (as in H_2 and O_2), the enhancement for NO is expected to be much greater than those for H_2 and O_2 for similar laser power densities. For systems (H_2/Ni , or O_2/Ni) where spin-dependent couplings dominate, the Landau-Zener estimate gives $\theta \approx 10^{-13} I (w/cm^2)$. For the H_2/Ni system, our results show an enhancement $\leq 20\%$ for $I = 10^8 w/cm^2$. For the O_2/Ni and similar systems, enhancement is observed only for $I \geq 10^{13} w/cm^2$, indicating that this class of systems is not likely to be a good candidate for laser/SMF predissociation. However the SMF in this case may be viewed as an agent to sustain

bound states in the presence of the laser, or to block reaction pathways involving the dissociated species. Hence it may also be regarded as a catalyst for excited bound-state reactions (when the laser is used as the excitation mechanism). For the NO/Ni,Co system, we estimate $\theta \approx 10^{-9} I (w/cm^2)$, and our results show that for enhancement of ~20%, one may have a much lower $I (\approx 10^5 w/cm^2)$. For $I \approx 10^8 w/cm^2$, the enhancement for the NO/Ni,Co system may reach 40%.

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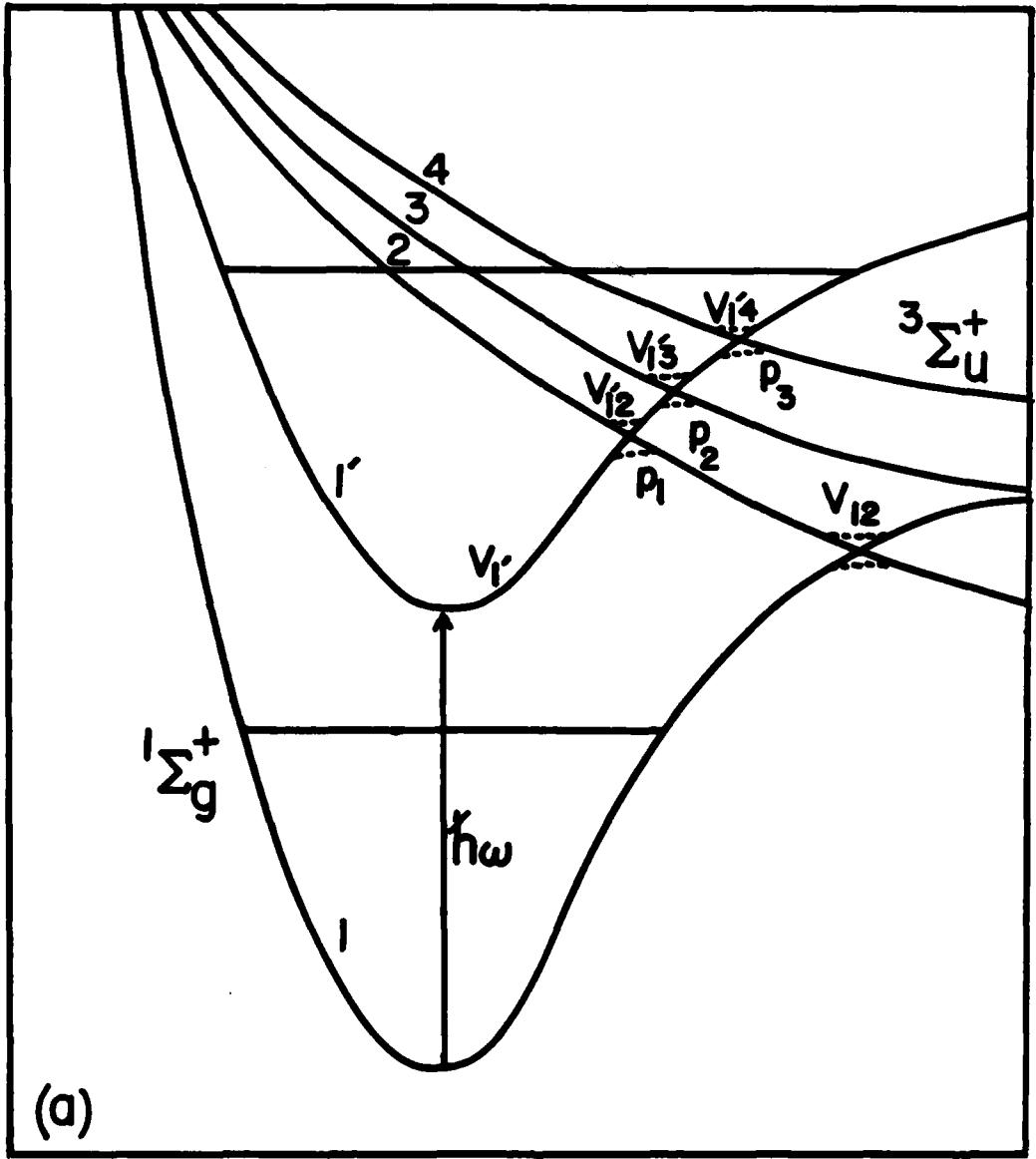
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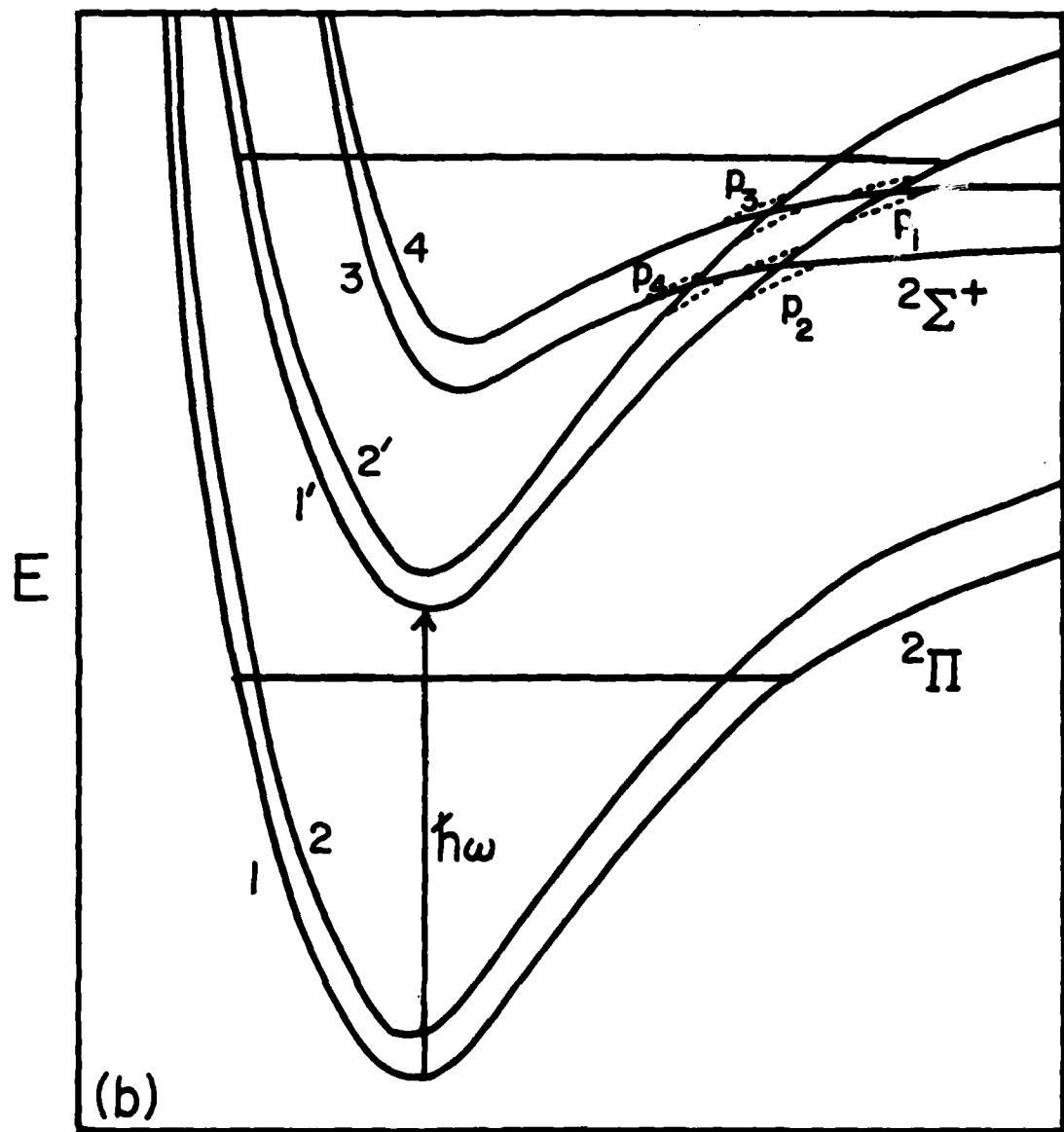
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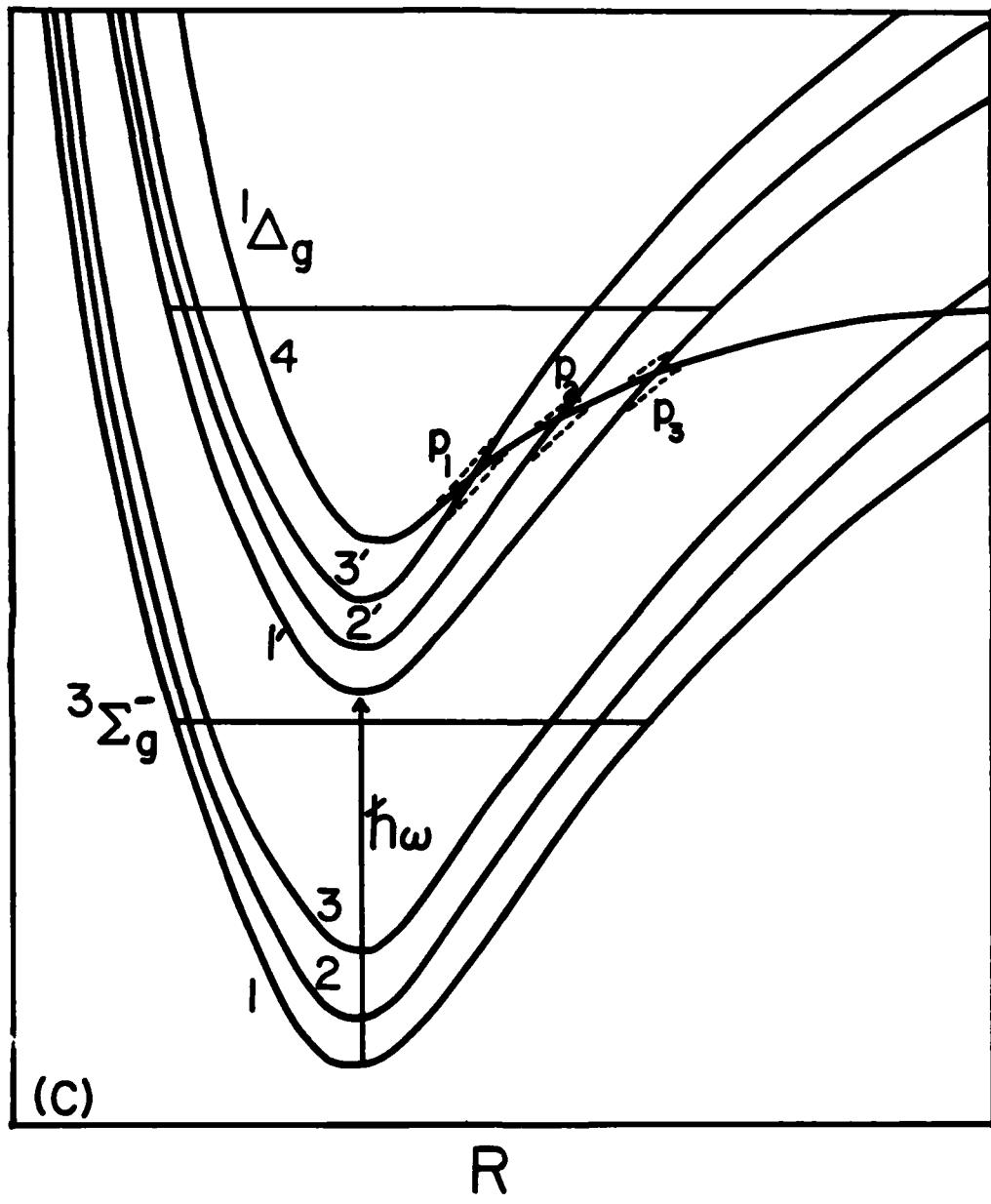
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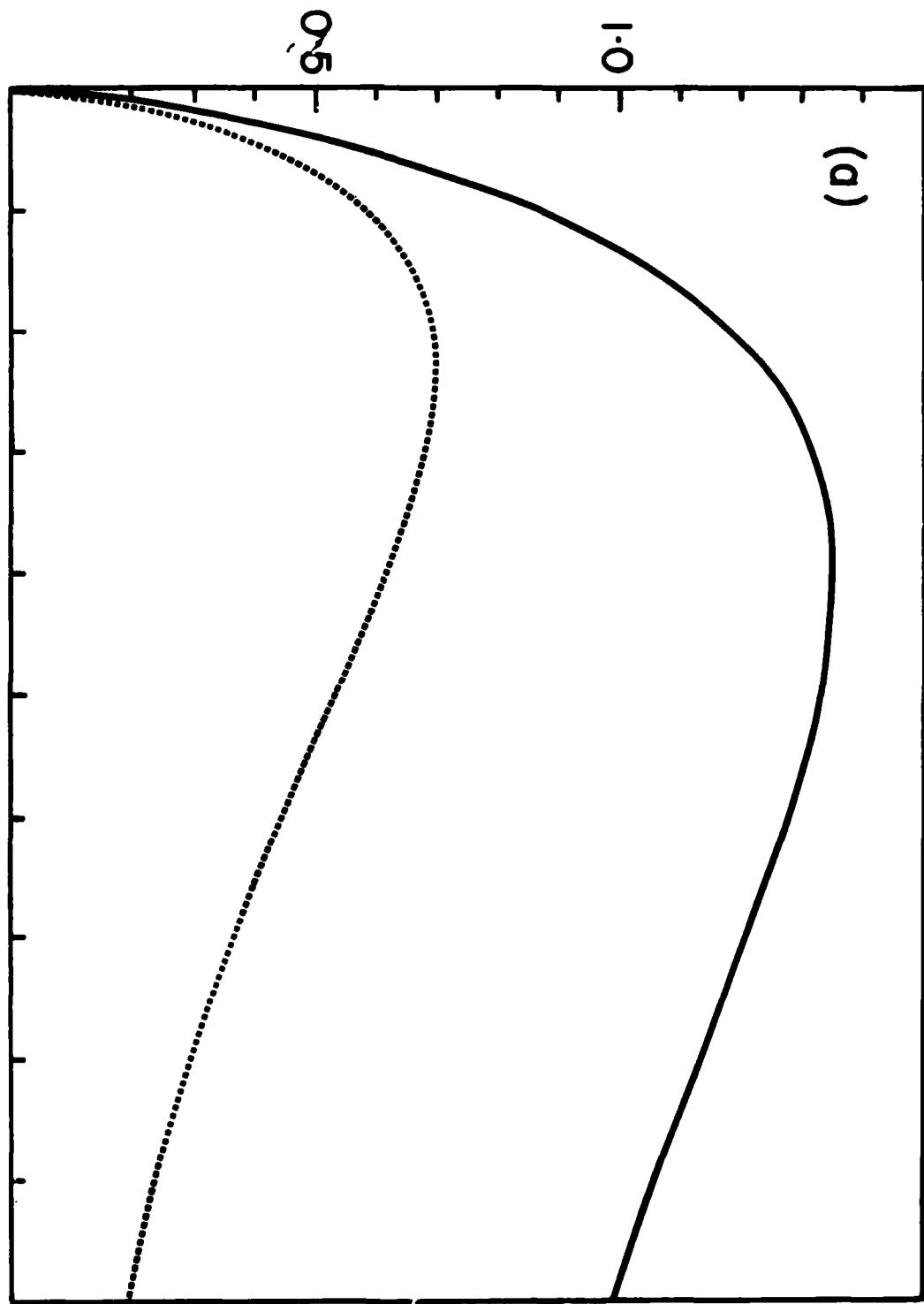
Fig. 1. Schematic drawings representing crossings among the field-dressed and the SMF-split electronic levels of (a) H_2 , N_2 , etc/metal, (b) NO/metal and (c) O_2 /metal-systems respectively. The laser-dressed states are designated by primed numbers. ω is the frequency of the laser photon. The V in (a) designate the vibrational quasibound wells. The p designate the local transition probabilities. The H_2 , N_2 /metal system has four, and both of the NO/metal and O_2 /metal-systems have nine such vibrational quasibound wells (see text).

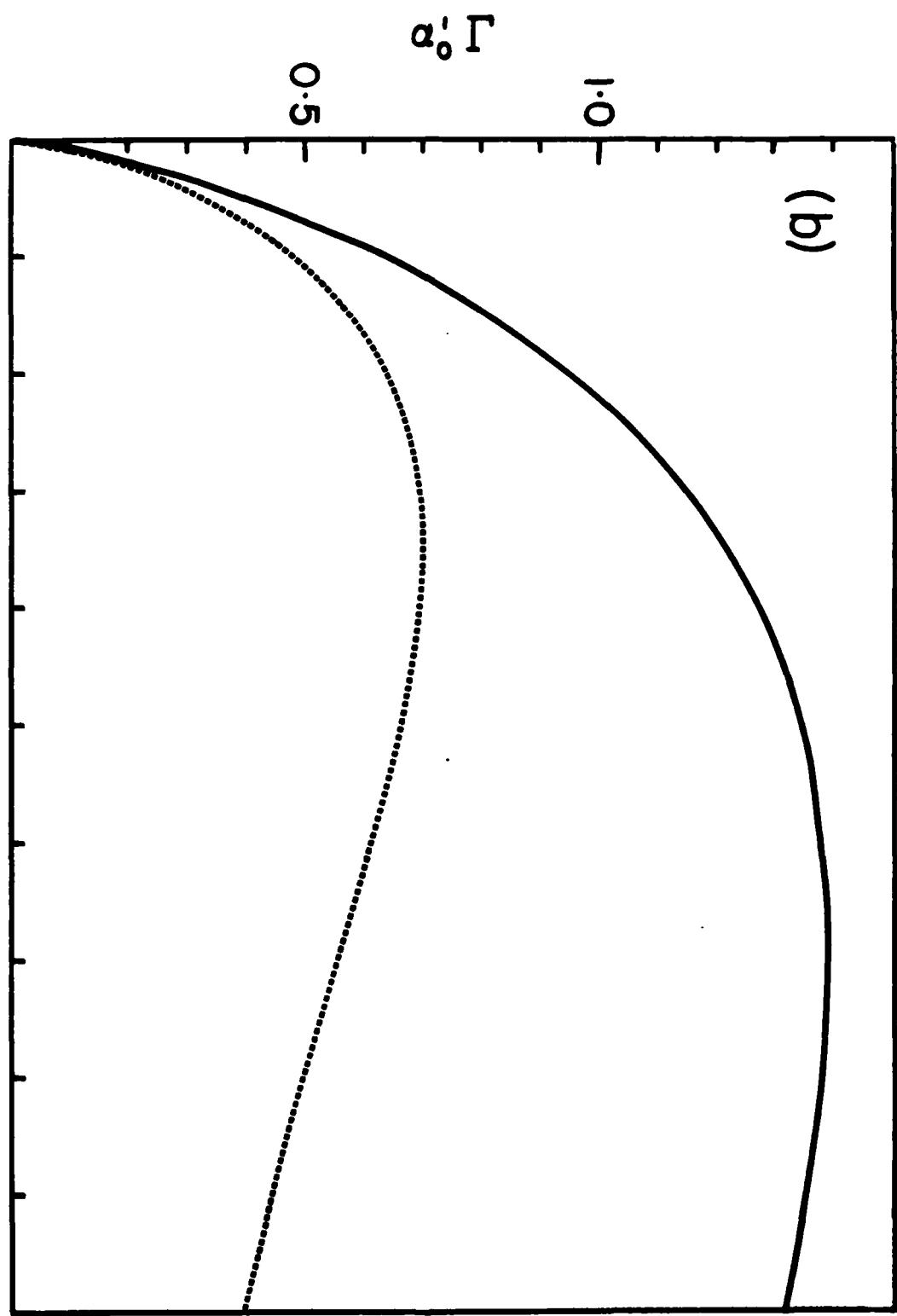
Fig. 2. Relative predissociation rates of adsorbed species:
_____, surface/laser; -----, gas-phase/laser.
 θ is proportional to the laser intensity. (a) H_2 ,
 N_2 etc/metal, (b) NO/metal, (c) O_2 /metal.

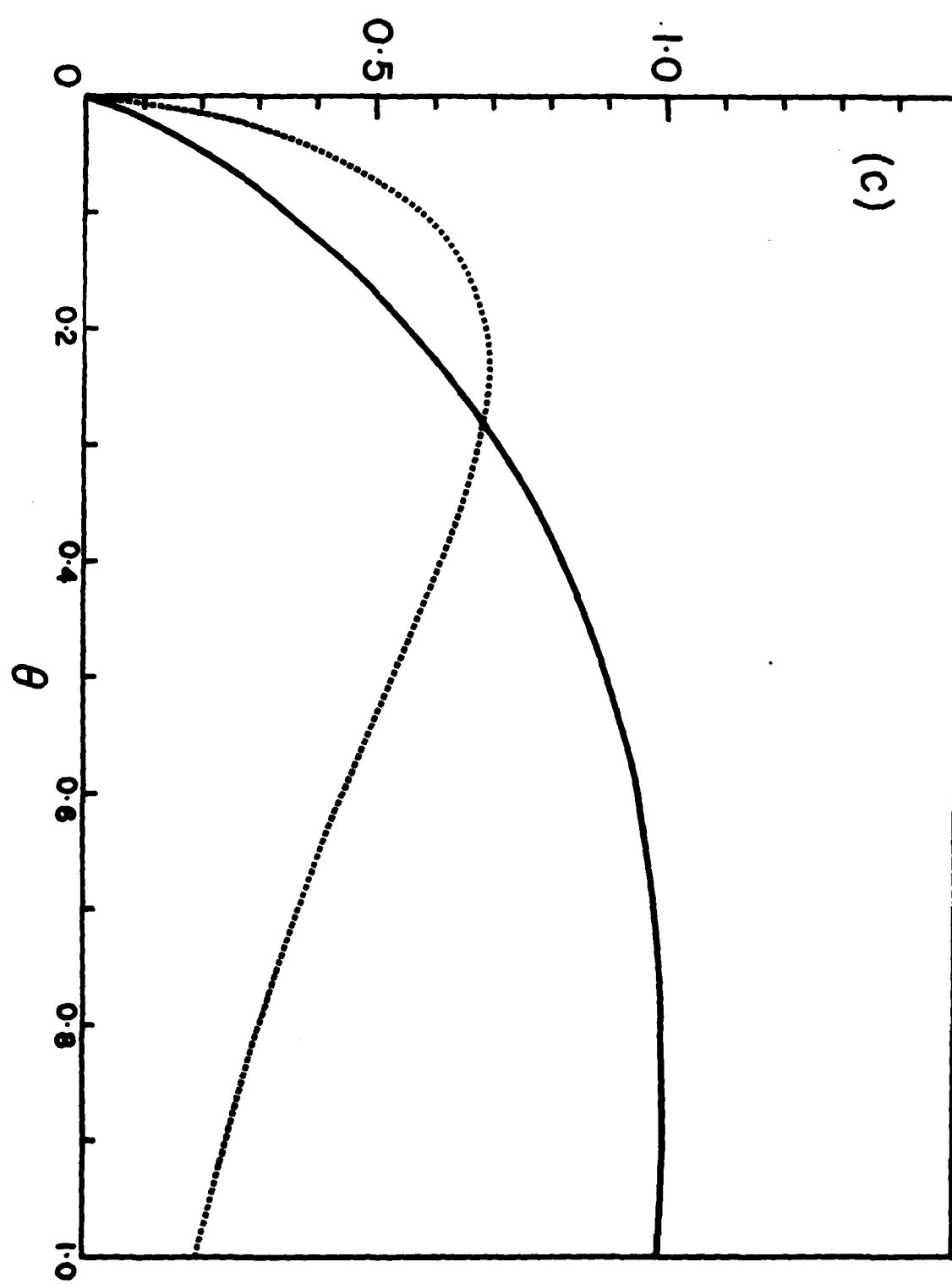












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